

Restriction Requirement under 35 U.S.C. 121

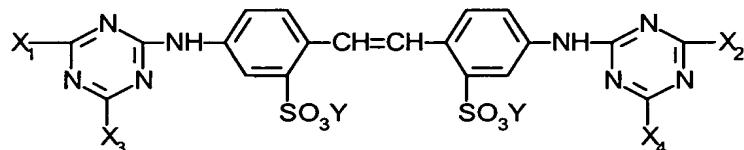
Applicants acknowledge election, but without traverse, of Group I of the Office Action, which includes Claims 1-11 drawn to diaminostilbenes. Applicants reserve the right to file divisional applications to the non-elected subject matter.

Rejection under 35 U.S.C. 103

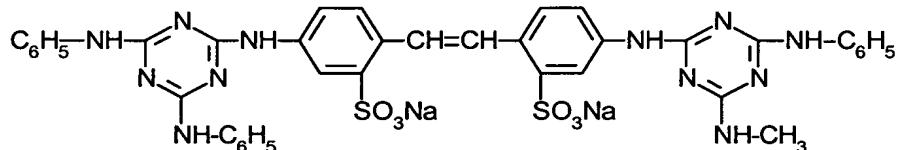
Claims 1-11 stand rejected under 35 U.S.C. 103(a) as being unpatentable over U.S. Patents 3,177,207 ("Siegel et al"), 3,655,574 ("Frishkorn et al"), or 3,895,009 ("Fringeli et al"). Applicants respectfully traverse.

A. Siegel et al.

Siegel et al discloses asymmetrically substituted bis-triazinylaminostilbene optical bleaching agents having the formula



in which X_1 , X_2 , and X_3 are various aromatic amino groups; X_4 is NH_2 or various aliphatic or araliphatic amino groups; and Y is hydrogen or an inorganic or organic cation. E.g., column 1, lines 11-36. [The more specific optical bleaching agent of Example 1, which is represented by the formula



is referred to in the Office Action at page 3.] Siegel et al teaches that the disclosed compounds can be prepared by several methods, including a three-stage process in which, **first**, cyanuric chloride (a trichloro-substituted heterocycle) is condensed at 0 to 10°C with 4,4'-diaminostilbene-2,2'-disulfonic acid in a 2:1 molar ratio; **second**, two of the chlorine atoms of the tetrachloro compound obtained in the first step are replaced with aromatic amino groups at 10 to 40°C; and **third** (in a step specifically designed to obtain an asymmetric substitution pattern), one of the two remaining chlorine atoms is replaced with an aromatic amino group and the other remaining chlorine atom is replaced with an NH_2 or an aliphatic or araliphatic amino group at 70

to 98°C. E.g., column 1, line 51, through column 2, line 11. More specific details of the reaction sequence are provided only in the examples, which teach that the third step was carried out by adding the methylamine and aniline mixture to the previously prepared dichloro compound and only then heating the reaction mixture to boiling. See Example 1 at column 4, lines 26-35.

Applicants, in contrast, claim a process for preparing diaminostilbenes of formula (I) (see Claim 13 above) by reaction of dichloro-substituted compounds of formula (IV) with various amines of formula (V) in an aqueous reaction medium under specific temperature conditions not taught by Siegel et al. In particular, Applicants' claimed process requires preparing a pre-heated aqueous reaction medium to which the dichloro-substituted compound of formula (IV) is added only after the reaction medium has been brought to a temperature at least 20°C higher than the temperature of the dichloro-substituted compound. Because the amine and any optional acid-trapping agent can be introduced before, during, or after the dichloro-substituted compound is added, the dichloro-substituted compound is subjected to elevated temperatures for a much shorter time than is possible for the process disclosed in Siegel et al (which necessarily requires exposing the dichloro-substituted compound to a heating process). The significance of this difference between Applicants' claimed process and the process disclosed in Siegel et al is shown by the comparative experiments described in the attached Declaration under 37 C.F.R. 1.132 of Rolf Brockmann, one of the inventors.

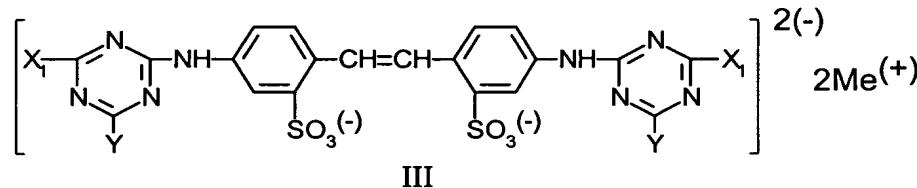
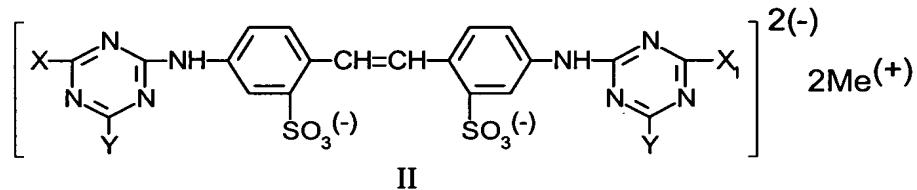
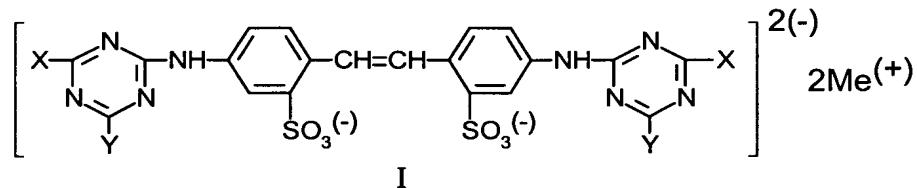
In Comparative Experiment A of the Declaration, Example 1 of Siegel et al was carried out except for using diethanolamine instead of a mixture of methylamine and aniline. In step (c) of the comparative experiment, diethanolamine was added to the reaction mixture of step (b), which contained the dichloro-substituted diamino-stilbene, and the reaction mixture was then heated to boiling. In Experiment B according to the present invention, the diethanolamine was dissolved in water and heated to 60°C, after which the reaction mixture of step (b) – which had been heated only to 35°C – was added to the aqueous ethanolamine. Chromatographic analysis of the respective products showed that Comparative Experiment A produced a yield of 83.67%, whereas Experiment B according to the invention produced a yield of 89.96%. The increased yield of 6.29% corresponds to a 7.5% increase relative to

the amount of product obtained in Comparative Experiment A. Nothing in Siegel et al suggests that controlling the temperatures in the manner specified by Applicants would result in improved yields. Such increased yields, however, not only represent a cleaner process but also can lead to reduced manufacturing costs.

In the absence of any suggestion of the control of temperature specified by Applicants, especially in view of the improved yields that Applicants have obtained, Applicants respectfully submit that Siegel et al does not render their claimed invention obvious.

B. Frishkorn et al.

Frishkorn et al discloses a process for preparing mixtures of symmetrical and asymmetrical bis-triazinylaminostilbene-2,2'-disulfonic acids having the formulas



in which X is an anilino group, X₁ is a morpholyl group, Y is a morpholyl or diethanolamino group, and Me⁺ is a hydrogen ion, metal ion, ammonium ion, or quaternary ammonium ion, in a multi-step process by, **first**, forming a tetrachloro-substituted condensation product of one equivalent of a metal salt of 4,4'-diamino-stilbene-2,2'-disulfonic acid and two equivalents of cyanuric chloride (see formula IV at column 2 of Frishkorn et al); **second**, condensing the initial condensation product at 10 to 40°C with two equivalents of amines of the formulas HX and HX₁; and **third**, condensing the resultant dichloro-substituted intermediate at temperatures no

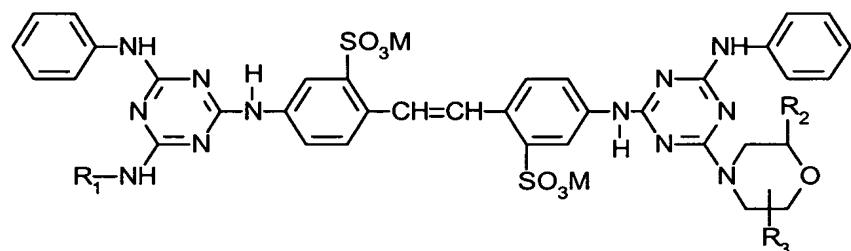
greater than 100°C with at least two equivalents of an amine of the formula HY; and **fourth**, isolating the product as the free acid by acidification. E.g., column 1, line 40, through column 2, line 23. [That is, the amino group asymmetry is introduced in the first reaction with amines, rather than in the second reaction with amines as taught by Siegel et al.] Although Frishkorn et al teaches that single components can be prepared for subsequent blending (e.g., column 2, lines 66-72), the clear emphasis of the reference is on preparing mixtures (e.g., column 1 and column 2, lines 70-72). None of the examples – including Example 1 cited in the Office Action – illustrates the preparation of individual compounds.

However, even if Frishkorn et al is read broadly to disclose the preparation of the individual compounds, Frishkorn et al – just as Siegel et al – discloses a process in which the dichloro-substituted intermediate is first mixed with amines and then heated and thus does not suggest the addition of a dichloro-substituted intermediate to a pre-heated reaction medium as required by Applicants.

Therefore, for essentially the same reasons as discussed above for Siegel et al, Applicants respectfully submit that Frishkorn et al does not render their claimed invention obvious.

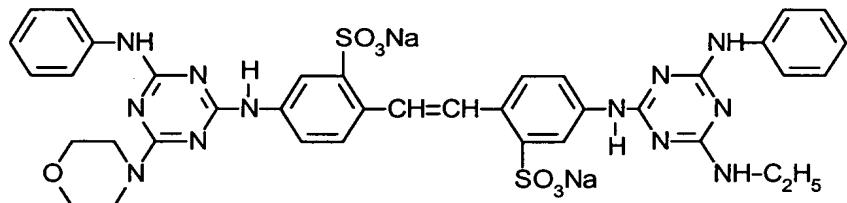
C. Fringeli et al.

Fringeli et al discloses 4,4'-bis-triazinylaminostilbene-2,2'-disulfonic acid derivatives having the formula



in which R₁ is C₁-C₄ alkyl, R₂ and R₃ are independently hydrogen or methyl, and M is a hydrogen, alkali metal, or optionally substituted ammonium cation. See formula (1) at column 1, line 35 et seq. One of the preparative methods described in Fringeli et al is a stepwise process in which the same basic tetrachloro-substituted starting material as disclosed in Siegel et al and Frishkorn et al is condensed with two molar equivalents of aniline and then with an excess of a mixture of morpholine (or a ring-

methyl derivative) and C₁-C₄ alkylamines. E.g., column 3, lines 25-36. The Office Action at page 5 specifically refers to the compound having the formula



for which the preparation was described in Example 5 (column 9). However, the final condensation step is carried out by mixing the dichloro-substituted intermediate with ethylamine and morpholine at 35 to 40°C, after which the reaction mixture was raised to a temperature of 85 to 90°C. See column 9, particularly at lines 31-33 and 40-42. Thus, Fringeli et al – just as Frishkorn et al and Siegel et al – does not suggest the addition of a dichloro-substituted intermediate to a pre-heated reaction medium as required by Applicants.

Therefore, for essentially the same reasons as discussed above for Frishkorn et al and Siegel et al, Applicants respectfully submit that Fringeli et al does not render their claimed invention obvious.

In view of the preceding amendments and remarks, allowance of the claims is respectfully requested.

Respectfully submitted,

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IN THE UNITED STATES PATENTS AND
TRADEMARKS OFFICE

APPLICANT: Ulrich Feldhues et al.
SERIAL NO.: 09/022,337
FILED: February 11, 1998
FOR: PROCESS FOR THE PREPARATION OF
SUBSTITUTED 4,4'-DIAMINOSTILBENE-2,2'-
DISULPHONIC ACIDS
GROUP ART UNIT: 1764
EXAMINER: A. Razgunas

DECLARATION UNDER 37 CFR 1.132

I, Rolf Brockmann declare:

1. That I am a German citizen and I reside at Piddelbornstr. 16, 51469 Bergisch-Gladbach, Germany;
2. That I am a chemist having graduated with the degree of Doctor rer. nat. at the Universität Gesamthochschule Paderborn, Germany in 1984;
3. That I have experience on the field of preparing optical brighteners since 1992;
4. That I am one of the inventors of U.S. Patent Application Serial No. 09/022,337;
5. That I am familiar with the subject matter of and have read the Office Action mailed March 3, 1999, and that the following experiments were carried out under my supervision:

Experiment A (comparative example)

This experiment was carried out according to Example 1 of U.S. Patent 3,177,207 ("Siegel et al") except that instead of a mixture of methyl amine and aniline, approximately the corresponding molar amount of diethanolamine was used in the third condensation stage.

- (a) 816 ml of deionized water was cooled to 2 °C. First 200 g of ice and then a solution of 50 g cyanuric chloride in 600 g of acetone were added. To the resulting suspension was added at 3-5 °C a neutral solution of 58 g of the sodium salt of 4,4'-diaminostilbene-2,2'-disulfonic acid in 476.3 g of water. pH value 7 was reached by adding sodium carbonate solution (15 %).

- (b) 26 g aniline were added within 5 minutes and the hydrochloric acid formed was neutralized with sodium carbonate solution until the pH value 7 was reached. The reaction mixture was heated to 35 °C.
- (c) 68.5 g of diethanolamine in the form of an 80.5% aqueous solution were added and the mixture was boiled under reflux (68 °C). The acetone was distilled off and the remaining solution was stirred for 4 hours at 98 °C.
- (d) The pH value was adjusted to 5 with hydrochloric acid. The precipitate was filtered off with suction while hot, washed with water and analyzed by high-pressure liquid chromatography.

Experiment B

This experiment was carried out according to Application Serial No. 09/022,337 (present invention).

Steps (a), (b), and (d) were conducted as described above. Step (c) was carried out as follows:

- (c) 68.5 g of diethanolamine were dissolved in water. The solution was heated to 60 °C and the reaction mixture of step (b) was added. The acetone was distilled off and the remaining solution stirred for 4 hours at 98 °C.

Result

The products of both experiments were analyzed by chromatography (HPLC) to determine the yield of the desired optical brightener. Experiment A produced an 83.67 % yield, whereas Experiment B produced an 89.96 % yield.

Conclusion

The process for the preparation of substituted 4,4'-diaminostilbene-2,2'-disulfonic acids according to Application Serial No. 09/022,337 (present invention) leads to a remarkably improved yield compared to the process according to the Siegel et al patent. Experiment B of the invention shows an increase in yield of 6.29 percent over comparative Experiment A.

The undersigned declarant declares further that all statements made herein of his own knowledge are true and that all statements made on information and belief are believed to be true; and further that these statements were made with the knowledge that wilful false statements and the like so made are punishable by fine or imprisonment, or both, under Section 1001 of Title 18 of the United States Code and that such wilful false statements may jeopardize the validity of the application or any patent issuing thereon.

Signed at Leverkusen, Germany, this 27 day of July, 1999.



ROLF BROCKMANN